

A study of the gamma rays of Pb^{206} from the decay of Bi^{206} by the method of external conversion in uranium

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With 6 figures in the text

Introduction

The Pb^{206} nucleus differs from the double magic Pb^{208} by two neutron holes. Pryce (1952) was able to predict the level structure of this nucleus on the basis of the shell model and the experimentally determined levels in Pb^{207} which is a single particle (hole) case.

The levels in Pb^{206} were studied by Alburger and Pryce (1954) in a detailed investigation of the decay of K -capturing Bi^{206} . Energies, spins and parities were established for ten excited states. Two less certain levels were also indicated with probable assignments. These experiments consisted of intensity and energy measurements of conversion lines in a double focusing spectrometer and studies of electron-electron and electron-gamma coincidences. A 145 microsecond isomeric state was discovered. The suggested decay scheme includes 28 transitions and shows satisfactory agreement with the theoretical predictions.

In some cases Alburger and Pryce could determine the multipolarities of the transitions from the K/L ratios of the observed conversion lines. From a scintillation spectrum, relative gamma ray intensities were estimated for a few cases, supporting the multipolarity assignments. Transition intensities were then deduced from the relative conversion line intensities and the theoretical conversion coefficients of Rose *et al.* (1951). In order to arrive at the best intensity balance Alburger and Pryce had to assume large $E2$ admixtures for some $M1$ transitions. The percentage of the $E2$ contributions varied in some cases between 20% and 50%.

The careful measurements of Alburger and Pryce resulted in the construction of a very complex decay scheme for Pb^{206} which seems to be well founded. For certain reasons it was nevertheless considered profitable to make a more detailed investigation of the gamma ray intensities than had been done by Alburger and Pryce.

Firstly, a scintillation spectrum, containing all the gamma rays found in Pb^{206} , can at most be used to ascertain that no very large deviations from the gamma ray intensities, as inferred from the conversion lines and the theoretical conversion coefficients, occur for the strongest transitions. No finer details can be expected from a resolution of 8–10% in such a case.

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Secondly, a thorough knowledge of the gamma ray intensities would give an important check on the decay scheme. In particular, it should be noticed that multipolarity determination on the basis of K/L ratios of conversion lines often fails for high energies and heavy nuclei. Here experimentally determined conversion coefficients would help very much.

Thirdly, according to Hultberg and Novakov (1957) photoelectron lines can now be analyzed in a more general way than has been possible before. From the point of view of the method the complex decay of Pb²⁰⁶ may serve the purpose of demonstrating the possibilities of estimating gamma ray intensities from a careful study of photolines.

As was done above, the following convention will be adopted throughout the text:

Electron lines, arising from the *internal* conversion of gamma rays, will be called *conversion lines*.

Electron lines, arising from the *external* conversion of gamma rays in thin foils, will be termed *photolines*.

Experiments

One of the difficulties with external conversion work is the requirement of rather large activities, often of the order of 100 times that necessary for internal conversion. This fact may even limit the number of isotopes which can successfully be subjected to this kind of investigation. In the case of Bi²⁰⁶ sufficient activity could be produced by bombarding metallic lead with deuterons in the cyclotron of the Nobel institute according to the reaction Pb²⁰⁶ ($d, 2n$) Bi²⁰⁶. In natural lead only 24% contributes to the formation of Bi²⁰⁶. In order to increase the yield, radiogenic lead was used because of its favourable isotopic constitution: 88% Pb²⁰⁶, 9% Pb²⁰⁷ and 3% Pb²⁰⁸. The energy of the bombarding deuterons was 16 MeV. Here, according to Alburger and Pryce, the production of longer-lived activities, e.g. Bi²⁰⁵, is practically eliminated. The irradiation lasted for 35 hours with an average deuteron beam of 50 microamps. After the bombardment a Pb layer 0.1–0.2 mm thick was scraped off. The dose rate from the lead activity was about 1 r/hour at 75 cm before the beginning of the chemical separation.

Apart from the question of self-absorption inside the source, it is generally not essential in external conversion work that the source be particularly thin. Also, for the sake of saving activity, a simple chemical separation of the active Bi from the bulk of the inactive lead was considered satisfactory. The irradiated lead was dissolved in hot nitric acid and the inactive lead precipitated as chloride. The Bi activity then remained in the liquid phase and the precipitate was removed by centrifuging. Addition of HCl and centrifugation were repeated several times. The volume of the solution was then decreased to a few drops by gentle heating in a water bath. The Al source holder (Fig. 1) for the spectrometer was filled dropwise with the active solution with subsequent evaporation to dryness. The source was cylindrical with a radius of 2.5 mm and a thickness of 1 mm. The dose rate was about 1 r/hour at 50 cm after this preparation.

The plane front side of the Al holder was covered with a circular uranium foil having a radius of 7 mm and a surface thickness of 3.0 mg/cm². Fig. 1 shows the experimental arrangement schematically.

Because of the great importance of the converters in experiments of this type, something should be said about their preparation and properties.

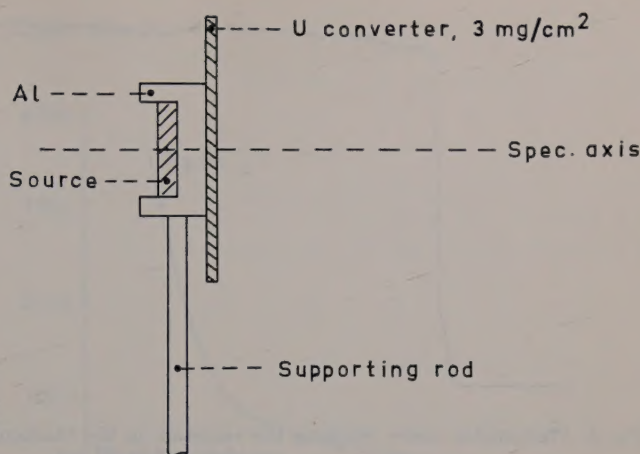


Fig. 1. Schematic view of experimental arrangement. Dotted line represents spectrometer axis.

The painting technique of Glover and Borrell (1955) was used for preparing the uranium foils. It is advisable to prepare an area which is larger than the size needed, in order to avoid edge effects. 0.1 mm aluminium has proved to be a suitable backing. The painting solutions used were mixed from the proportions

	I	II
Uranyl nitrate, mg	50	200
Ethyl alcohol, ml	2.5	3
4 % collodion, ml	10	3
Acetone, ml	7.5	10

Solution I is comparatively thin, giving for each painting 0.015 mg U per cm^2 , while solution II gave about 0.040 mg U per cm^2 .

First of all the backing should be carefully cleaned. The painting can be done with an ordinary camel hair artist brush. When putting on a layer, enough solution should be spread over the surface so that the uranium salt can distribute uniformly over the whole area before the solvent has evaporated. The drying may require one or two minutes and may be speeded up by gentle heating. In order to convert the uranyl nitrate into uranium oxide, the foil is then heated to 500–550°C, preferably in a closed oven, for about 10 minutes. The temperature and the heating time in the oven proved to be very important for obtaining good uniformity. After taking the foil out of the oven it should be carefully polished with a piece of cloth in order to remove any loose particles from the surface. The next layer may then be painted, subsequently dried, heated in the oven and so on until the required surface thickness has been reached. By this procedure 5 uranium foils were prepared according to the following table:

Foil no.	Surface thickness, mg/cm^2	No. of layers	Total area, cm^2
1	0.50	35	63
2	1.01	70	63
3	1.50	100	63
4	2.00	123	63
5	3.01	145	63

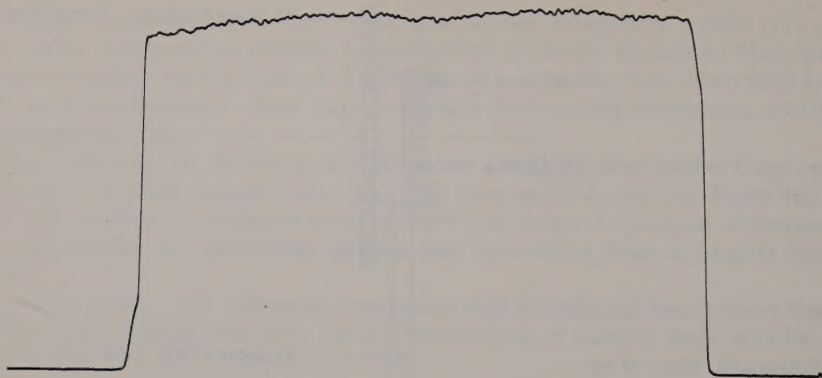


Fig. 2. Photometer curve showing the variation in the blackening of an X-ray film, placed in contact with a U foil.

The thin uranium solution was used except for the last mg/cm^2 of foil no. 5, which was painted with the thicker solution. The surface thicknesses have been determined by weighing. It can be seen from the table that the amount of U put on for every layer is very constant and thus it was easy to obtain desired values of the thicknesses. It is consequently felt that the quoted surface densities are quite accurate.

In order to test the uniformity of the prepared U foils they were placed in direct contact with X-ray films, which were exposed to the alpha-activity in this way for periods ranging from 24 hours to about one week. After development the obtained blackenings were studied in a photometer. Fig. 2 shows a typical result. The mean variation in the homogeneity was found to be less than 2% over the whole foil areas. Using the photometer recordings small circular foils for use in external conversion work could be cut out having a homogeneity better than 1%.

The large double focusing spectrometer of 50 cm mean radius was used for the measurements. Care was taken to minimize the material necessary around the source in order to depress the inevitable Compton background. The Al holder with the activity was supported by a thin Al rod in such a way that the U foil held the ordinary source position in the spectrometer. Al-lined lead bricks were placed behind the spectrometer aperture to decrease the background.

In all spectrometer work it is of prime importance that the ratio between the peak height of the recorded line and the background be as high as possible. An experiment showed that this ratio may be considerably increased by simply decreasing the distance between source and converter as far as possible. Fig. 3 illustrates a specific case where the major part of the photoline background arises from electron production and scattering in the spectrometer walls and to a negligible extent in the source holder and converter. This is obvious from the approximate constancy of the background curve over a large variation of the source-converter distance, which should be reduced as far as possible from other reasons as well (see below). An Al shielding having a thickness of 1.5 mm between source and converter was considered suitable for suppressing the conversion electrons sufficiently.

Counts/min

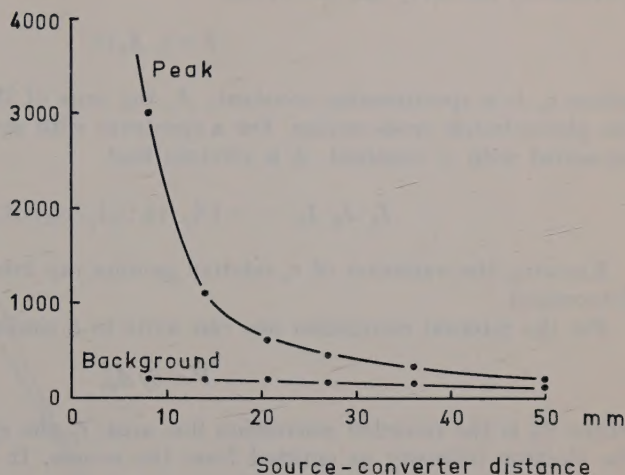


Fig. 3. The photoline peak and background counting rates plotted as functions of the source-converter distance. Recordings of the Cs 662 keV gamma ray.

The spectrometer was run in the conventional manner, i.e. the magnet current was varied in only one direction. A single GM-counter with a thin mylar window was used. It was operated with a gas mixture of argon and propane.

Four runs of the Pb^{206} gamma ray spectrum were made. Of these the first served as a check on the more prominent lines for orientation purposes, and in the fourth run the smaller peaks were measured separately. Backgrounds without the uranium foil were not taken because (a) the activity available did not permit of such extended measurements and (b) the flat portions of every line were very carefully recorded on both sides of the peak, thus permitting of a safe interpolation of the background under the peak.

Analysis of results

Fig. 4 shows a complete set of recorded photolines. It can be seen that in most cases the lines are completely resolved. The widths of the photolines at half maximum varied from 0.8% to 0.4% of the abscissa for quantum energies in the region 300–2000 keV, reflecting the variation in energy loss of the photoelectrons with energy. A constant resolution would have required the use of several foils with different thicknesses. For the present experiments this was not considered necessary.

For the energy calibration of the spectrometer, a curve similar to that of Fig. 3 in the paper by Hultberg and Hedgran (1956) was constructed, giving H_Q as a function of the spectrometer variable Ω . A period of 6.4 days, as quoted by Alburger and Pryce, was used for the decay corrections. As expected from the choice of the deuteron bombarding energy, no Bi^{205} lines could be detected.

From the measured photolines, as shown in Fig. 4, the intensities of the corresponding gamma rays can be inferred. Following Deutsch, Elliott and Evans (1944), the gamma ray intensity can be written

$$I = c_\gamma A_\gamma / \tau, \quad (1)$$

where c_γ is a spectrometer constant, A_γ the area of the recorded photoline and τ the photoelectric cross-section. For a spectrum with several gamma rays 1, 2, 3 ..., measured with c_γ constant, it is obvious that

$$I_1 : I_2 : I_3 : \dots = (A_\gamma / \tau)_1 : (A_\gamma / \tau)_2 : (A_\gamma / \tau)_3 \dots \quad (2)$$

Knowing the variation of τ , relative gamma ray intensities can consequently be determined.

For the internal conversion one can write in a similar manner

$$P = c_\beta A_\beta, \quad (3)$$

where A_β is the recorded conversion line area, c_β the spectrometer constant and P the electron intensity as emitted from the source. It follows that

$$P_1 : P_2 : \dots = (A_\beta)_1 : (A_\beta)_2 : \dots \quad (4)$$

Division of Eq. (4) by Eq. (2) gives for the conversion coefficients ϵ

$$\epsilon_1 : \epsilon_2 : \dots = \frac{P_1}{I_1} : \frac{P_2}{I_2} : \dots = \left(\frac{A_\beta}{A_\gamma} \cdot \tau \right)_1 : \left(\frac{A_\beta}{A_\gamma} \cdot \tau \right)_2 : \dots \quad (5)$$

Fixing one of the conversion coefficients, the rest can be obtained from Eq. (5).

For the definition of symbols and terminology used in the following text pertaining to the mechanism of the photoelectric emission, the reader is referred to the paper by Hultberg (1955).

According to the procedure described by Hultberg and Novakov (1957), appropriate values of the photoelectric cross-sections were obtained for the geometry of the experimental set-up. For three energies "dotted curves" of the type in Fig. 1 of the last-mentioned paper were integrated from 0° to 75° , the latter angle being the maximum angle of emission ϑ_m for the photoelectrons recorded in the spectrometer. A small uncertainty in ϑ_m (of the order of a few per cent) will cause a negligible change in the computed gamma ray intensities. The integration was carried out at three quantum energies: 411, 662 and 1332 keV. The photoelectric cross-sections obtained were then plotted versus energy, as shown in Fig. 5. Below 300 keV the extrapolation is uncertain, which is indicated by the dotted part of the lower curve of Fig. 5, where a comparison is made between the photoelectric cross-sections to be applied in the analysis of these experiments and the theoretical values for the total absorption in the K -shell. The difference for small energies is clearly seen to be large in spite of the fact that care has been taken to increase ϑ_m as much as possible by minimizing the source-converter distance. In external conversion work, ϑ_m is often of the order of 30° – 50° , in which case this difference will be much larger.

From Eq. (2) the relative gamma-ray intensities were computed, taking τ from the lower curve of Fig. 5. To obtain conversion coefficients, Eq. (5) was used, substituting

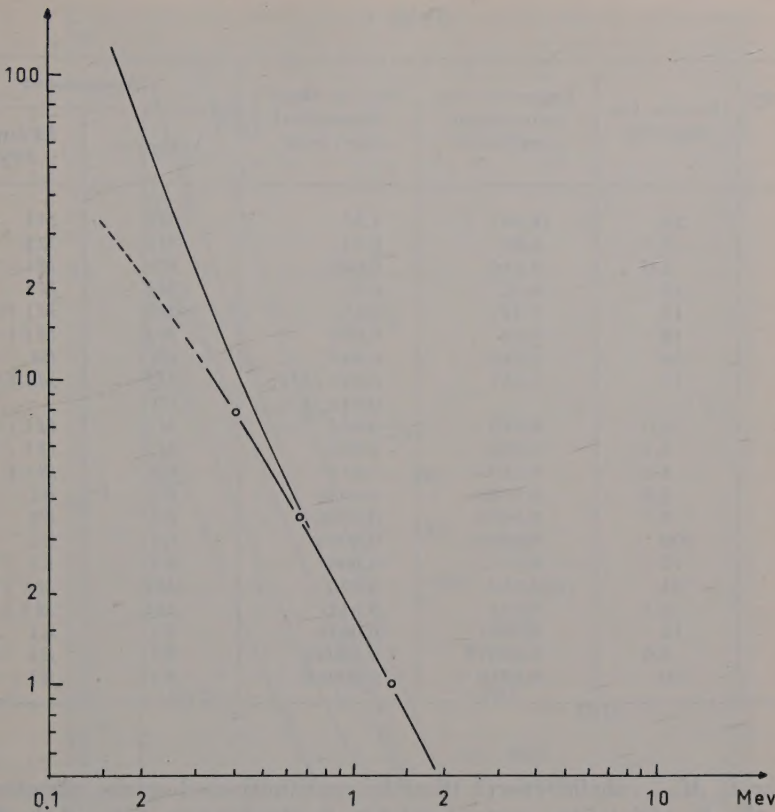


Fig. 5. Comparison between theoretical total absorption cross-sections for the K -shell (upper curve) and the photoelectric cross-sections to be applied to the analysis of the Pb^{206} photolines (lower curve).

the relative K -conversion intensities of Alburger and Pryce for A_β . Concerning normalization, see below.

Mean values of the relative gamma ray intensities and conversion coefficients thus found from the 4 runs are presented in Table 1 together with multipole assignments. For comparison the theoretical values of Sliv and Band (195) are also given. Our computed energy values agreed to within 0.3% with those of Alburger and Pryce and thus will not be presented.

In this connection an interesting feature should be noticed. In a few cases the K photoelectron lines were not resolved but more or less mixed with L photolines. Moreover, in the case of the 184 keV gamma ray only the L lines were observed due to recording difficulties. The conversion coefficients have nevertheless been computed even for these cases. Here the observation that the K/L ratio for photoelectrons is essentially constant within a large energy range has been used (Heitler, 1954). Unfortunately, accurate experimental values for this ratio are lacking; Heitler states only that about 80% of the photoelectric absorption takes place in the K -shell and it seems that nothing definite is known concerning the distribution of the remaining

Table 1.

Gamma ray energy (keV)	Gamma ray intensity	Experimental conversion coefficient	Sliv's & Band's theoretical conv. coeff.	Assignment	
				Author's	Alburger & Pryce
184	21	[1.05]	1.35	M1	M1
263	2.9	0.66	0.51	M1	M1
314	3.0	0.046	0.057	E2	M1
343	31	0.22	0.25	M1	M1
398	13	0.14	0.17	M1	M1 (+ E2)
497	18	0.08	0.091	M1	M1 (+ E2)
516	50	0.043	0.049	E3	E3
537	47	0.047	0.075 (M1)	M1 +	M1 (+ E2)
			0.018 (E2)	+ E2	
621	4.0	0.073	0.052	M1	M1 (+ E2)
632	4.5	0.050	0.050	M1	M1 (+ E2)
657	5.3	0.016	0.012	E2	M1 (+ E2)
740	3.6	0.0033	0.0036	E1	E1
754	2.7	0.0029	0.0035	E1	E2
803	100	0.0090	0.0082	E2	E2
880	72	0.0072	0.0065	E2	E2
895	21	0.017	0.020	M1	M1 (+ E2)
1019	8.1	0.014	0.014	M1	M1 (+ E2)
1099	12	0.0024	0.0018	E1	E1
1596	8.0	0.00073	0.00093	E1	E1
1720	30	0.0010	0.00082	E1	E1

20% of the L , M , ... shells, except that the contributions decrease monotonically. However, the K - and L -lines were completely resolved for the 516 and the 880 keV energies and so two values of the K/L ratio were obtained (around 5,5), assuming the directional dependence of the L photoelectrons to be such that practically no electrons are emitted at angles larger than ϑ_m (defined above by the experimental set-up). For energies above 600 keV this assumption seems to be in accordance with preliminary experiments on the directional dependence of L photoelectrons carried out by Hultberg and Novakov (unpublished). Thus it was in two cases possible to deduce the magnitudes of the components of the compound peaks without making any specific assumptions about the actual line shapes. It is evident that in external conversion more results may be reached than the actual resolution would seem to permit. However, far too little is as yet known about the angular dependence of L -shell photoelectrons and its energy variation to permit of a more extensive use. It is planned to continue the preliminary experiments already made on this problem in more detail.

The conversion coefficient of the 184 keV transition has been bracketed because it is considered uncertain. The energy is far from the lowest calibration point at 411 keV of the photoelectric cross-section curve (Fig. 5) and only the L -line was recorded. Moreover, it is for the moment impossible to tell how good the constancy of the K/L ratio at such small energies is and, in addition, intensity loss due to self-absorption inside the source becomes serious in this region. It is consequently impossible to give an accurate value of the gamma ray intensity for this line. Furthermore, according

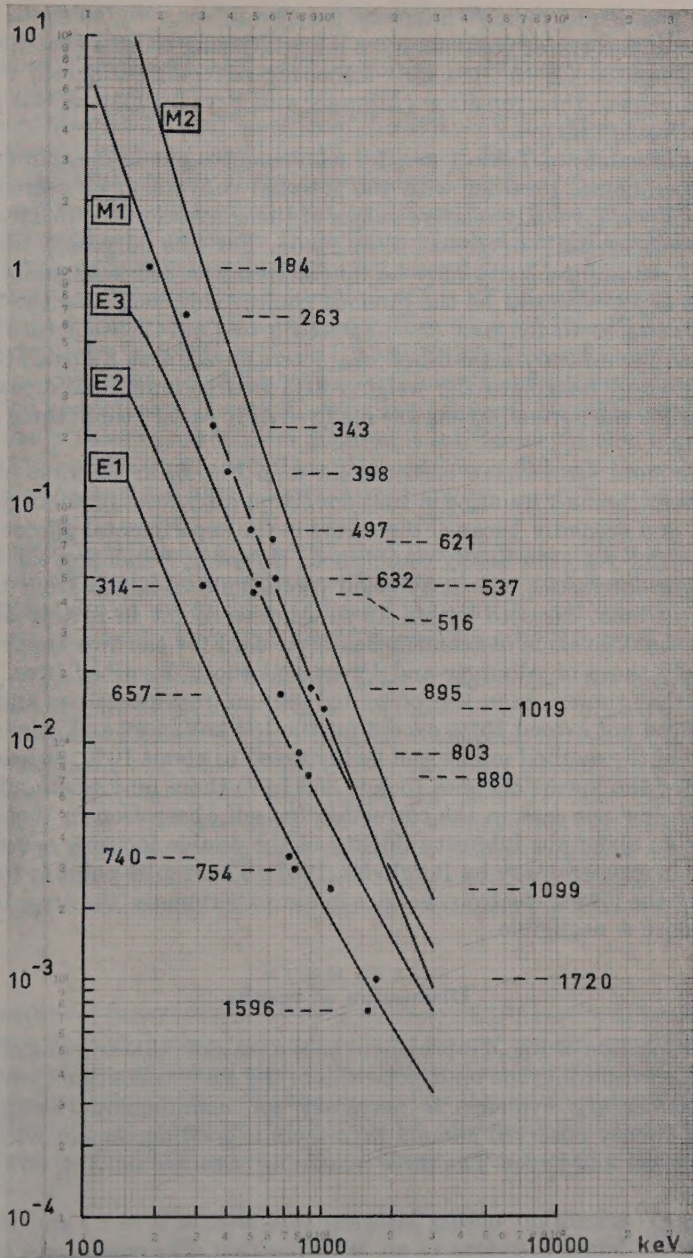


Fig. 6. Comparison between Sliv's and Band's theoretical conversion coefficients (full lines) for $Z = 82$ and experimental values for Pb^{206} (figures indicate gamma ray energies in keV).

to Alburger and Pryce their K -intensity may be in error by as much as 30%. In spite of these unfavourable circumstances it is gratifying to note that the conversion coefficient computed for the 184 keV transition nevertheless agrees well with the earlier assignment of this transition (Alburger and Pryce, 1954), as this is one of the more certain cases ($M1$).

In Fig. 6 the values of Table 1 are, for convenience, put in diagrammatical form. As these experiments, together with the internal conversion measurements of Alburger and Pryce, give only relative values of the conversion coefficients, a normalization to the theoretical curves has to be made. For this purpose a transition with definitely known multipolarity may be taken. Here the 803 keV gamma ray to the ground state of Pb²⁰⁶ would be the obvious choice. However, also the 880 keV ray from the third ($4+$) to the first ($2+$) excited levels and the isomeric 516 keV transition seem to have firmly established characters as $E1$ and $E3$ transitions, respectively. In order to distribute the weights attached to individual measurements as much as possible the normalization was made so as to satisfy these three gamma rays about equally.

No experimental errors have been indicated in Fig. 6, in the first place because no details about the uncertainties in the conversion intensities of Alburger and Pryce were known and secondly because the errors in the experimental photoelectric cross-sections have not yet been finally established. However, neglecting the errors in the conversion line intensities, the conversion coefficients obtained from the different runs agreed in most cases to within 15%. Another point in favour of the values presented is that the same instrument has been used for the two types of measurements, namely those of Alburger and Pryce and those described here. It is consequently felt that the errors in the presented gamma ray intensities and conversion coefficients need not exceed 15%, except for the 184 keV case, and it is believed that the accuracy of the method might well be increased to about 10%, at least in favourable cases. For the lowest energies (below 350 keV) there may be an additional uncertainty of a few per cent in the correction for self absorption in the source.

The question about the intensity fidelity of the double focusing spectrometer has been studied in a general way by Bergkvist (1957). The results point to the conclusion that, even for the lowest electron energies in our experiments, the error in the instrumental response is negligible.

Discussion of results

From an inspection of Fig. 6 it is evident that a considerable amount of information can be obtained regarding the multiplicities of the Pb²⁰⁶ transitions, even if, in some cases, complementary evidence is necessary for unambiguous assignments. For seventeen of twenty observed gamma rays there is good agreement with the assignments of Alburger and Pryce. The three remaining cases are the 314, 657 and 754 keV gamma rays.

The 314 keV transition occurs between the 1998 keV ($4+$) and the 1684 keV ($4+$) states and should therefore be an $M1$ transition. The conversion K/L ratio was not determined by Alburger and Pryce, probably because of the weakness of the L line. Our result would instead indicate an $E2$ transition.

In the case of the 657 keV gamma ray, which depopulates the same 1998 keV level, the K/L ratio was found to be 5.4. The experimental error is rather large and the limits are about 6.5 and 4.2. Our conversion coefficient would indicate an $E2$

transition in this case also. The theoretical K/L ratio for an electric quadrupole at the energy in question is 3.9 which is not too much outside the experimental error. However, in both of these cases the possibility of $M1 + E2$ with a strong $E2$ admixture cannot be excluded.

The third transition of 754 keV goes from the 3280 keV ($5 -$) level to the 2525 keV ($3 - ?$) state. Our assignment would be $E1$ instead of $E2$. It is interesting to notice that the 2525 keV state is one of the two doubtful levels.

It should perhaps be emphasized that the photolines from these three gamma rays were very weak and therefore no definite conclusions are possible.

For the 537 keV gamma ray, going from the second ($3 +$) to the first ($2 +$) excited states, Alburger and Pryce had to assume an $M1 + E2$ mixture in the ratio 5 to 3 in order to obtain a proper intensity balance in the decay scheme. From Fig. 6 in this paper the mixing ratio is found to be 1:1 which is in fair agreement with their requirement. Our conversion coefficient was here obtained from different runs which all showed good consistency. However, using the conversion coefficients of Sliv and Band the intensity balance of the term diagram is satisfactory within a few per cent, even if the 537 keV ray is a pure $M1$. This inconsistency is difficult to explain because both the conversion and the photoelectric intensities are carefully measured for this gamma ray.

In conclusion, it is interesting to notice that the comparison of the experimental values with the theoretical conversion coefficients of Sliv and Band, who take into account the finite size of the nucleus, indicates that the $M1$ transitions are, even at fairly high energies, much more pure than was believed by Alburger and Pryce.

Note added in proof: Very recently a new series of experiments has been performed at the Nobel Institute by one of the authors (S. H.) in cooperation with R. Stockendal. The purpose was the *absolute* determination of conversion coefficients by measuring external and internal conversion using the same source (or two identically prepared sources with known relative strengths). Such experiments were carried out on Cs-137 and Pb-205, 206. The preliminary results seem to be satisfactory. Particular attention was paid to the above mentioned 537 keV transition in Pb-206 with the hope of resolving the observed inconsistency discussed above. Details of these experiments will be published later in Arkiv för fysik:

S. Hultberg and R. Stockendal on Cs-137 and R. Stockendal and S. Hultberg on Pb-205, 206.

SUMMARY

Relative intensities of 20 gamma rays following the decay of Bi^{206} have been determined from a beta-spectrometric study of photoelectrons ejected in a 3 mg/cm^2 uranium converter. The instrumental resolution was 0.4 % at high energies. A new method of analysis of the recorded photolines has been applied. Together with the earlier measurements of the internal conversion by Alburger and Pryce, the gamma ray intensities found permitted a determination of relative conversion coefficients with an accuracy of about 15 %. In most cases the earlier multipolarity assignments were confirmed.

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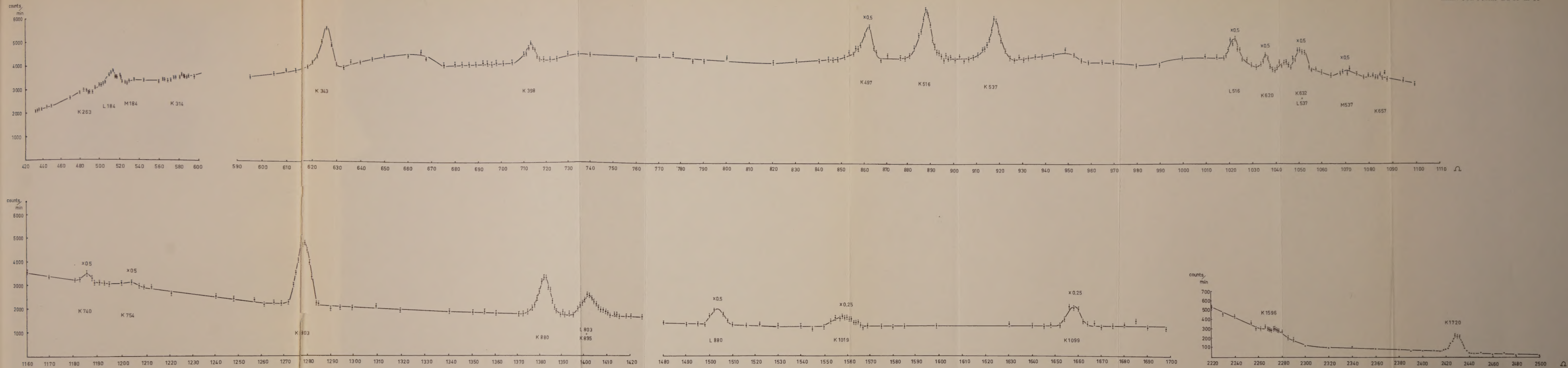


Fig. 4. A spectrometer recording of the photolines arising in a 3 mg/cm² U foil by the action of the Pb²⁰⁸ gamma rays. The indication $\times 0.5$ means that the observed counting rate will be obtained by multiplication with 0.5.

